REMARKS

Claim 2 is amended; claims 1-39 are pending in the application. Claims 1 and 7-10 stand rejected as obvious in view of *Dean*; claims 2-6 and 11-39 stand rejected as obvious in view of *Dean* in combination with either *Johnson* or *Herbst*. The amendment to claim 2 corrects a minor error for the purpose of clarification and presents no new matter. Further examination of the application as amended and reconsideration of the rejections are respectfully requested.

By way of background, applicant's invention as recited in claim 1 provides a method for supercritical conversion of high-boiling hydrocarbons (>1000°F) to low boiling hydrocarbons (<1000°F). The feed high-boiling hydrocarbons are supplied in a mixture with a solvent (boiling point <1000°F) at a minimum weight ratio of two-thirds solvent to no more than one-third feed hydrocarbons. The feed/solvent mixture is heated by admixture with hot particles, which need not be catalytic (see the specification at 22/12 through 23/13, especially 23/4-9), and maintained at a temperature and pressure that exceed the critical temperature and critical pressure of *the solvent/feed hydrocarbon mixture*. The presence of the solvent alters the mixture critical properties, generally by reducing the critical temperature relative to the high-boiling hydrocarbons in the feed and

increasing the critical pressure relative to both the feed and solvent hydrocarbons. See the specification at 37/9 through 41/2 and Figs. 8-14, especially 38/9-22. Converted low boiling hydrocarbons are recovered and separated from solids upon which coke is deposited.

Dean discloses a fluid catalytic cracking (FCC) process that employs a special ultrastable crystalline zeolite of high silica to alumina ratio that is said to increase light cycle oil formation, while reducing heavy cycle oil and coke make. The office action alleges, "Dean discloses an FCC process wherein a heavy oil feed having components boil above 1000°F [*sic*, 600 °F] is charged into a riser reactor to contact with a hot catalyst at a temperature equal to or above the feed pseudo critical temperature . . . Dean also discloses that the feed is mixed with light hydrocarbon. See col. 2, lines 16-38; col. 8, line 2 through col. 9, line 9; claims 1 and 10." The cited passages from *Dean* read as follows:

> <u>2/16-38:</u>
> "The catalytic cracking or conversion of heavy oil feeds comprising atmospheric tower bottoms vacuum gas oils plus resid, residual oil, reduced crudes, and topped crudes, shale oil, coal liquefaction oil products and tar sands oil products all of which comprise components boiling above 1000°F. or 1050°F. are best *catalytically converted in a highly vaporized-atomized contact phase of the oil feed* with select fluid catalyst particles of a composition herein identified. The heat necessary to *substantially completely vaporize-atomize all of the high boiling oil feed* is contributed by a relatively high regenerator temperature operation of at least about 1350°F. and preferably above 1400°F. completed in the substantial absence of hydro-thermal damage to the catalyst. The more conventional zeolite cracking catalysts comprising relatively large amounts of one or more rare-earth exchanged faujasite zeolites are susceptible to considerable hydro-thermal deactivation under high temperature regeneration conditions above about 1400°F.

and required to remove relatively large amounts of hydrocarbonaceous materials deposited during residual oil cracking operations." [*Emphasis added.*]

8/2 – 9/9:

". . . equal to the feed pseudo-critical temperature. Thus the catalyst regeneration temperature will increase above 1350°F. during combustion of deposited carbonaceous material as the residual oil feed Conradson carbon value increases even though the ultrastable zeolite catalyst composition employed in the conversion process of this invention contributes to a reduction in coke make. Thus when processing vacuum gas oils comprising the resid portion of the crude oil, regeneration temperatures above 1350°F. and up to as high as 1600°F. or more can be experienced as carbonaceous deposits increase in response to the feed Conradson carbon content.

"The catalytic conversion operation of this invention is preferably one of relatively short vaporized hydrocarbon contact with the special catalyst composition comprising from 20 to 80 wt % of ultrastable crystalline zeolite as a dispersed catalyst phase in a riser contact zone wherein the hydrocarbon residence time in contact with catalyst particles can be restricted to within the range of 0.5 to 5 seconds and more usually in the range of 1 to 3 seconds. *This dispersed catalyst phase-vaporized hydrocarbon contact may be implemented in substantial measure by the use of an atomizing diluent material with the high boiling hydrocarbon feed. Diluent materials suitable for this purpose include steam, CO2, light normally gaseous hydrocarbons comprising C3 minus material or a combination thereof <u>in an amount which will reduce the high boiling feed partial pressure</u> and achieve desired atomized-vaporized dispersion contact of hydrocarbon feed with high temperature catalyst particles. Atomization of the feed may be substantially implemented by use of appropriate spray nozzles. Thus the operating parameters to achieve an optomized [sic] contact between feed and catalyst particles also include feed exit velocities in excess of 10 feet per second to achieve atomized spraying of the feed with or without diluent material across a riser reactor cross section for intimate contact with hot catalyst particles charged thereto.*

"The above identified operating parameters are intended to also accelerate the mixture relatively uniformly within the feed vaporization section of a riser reactor in a minimum time frame and thus enhance rapid heat transfer from hot catalyst particles to charged feed preferably atomized and thus prevent localized enhanced catalyst to oil ratios contributing to a dense catalyst bed phase. That is, the operating conditions and methods for implementing are selected to ensure a relatively dilute phase suspension contact between catalyst particles and atomized oil feed for vaporized conversion transfer through a riser conversion zone. Such dilute catalyst phase operations include catalyst particle concentrations in the range of 2 to 10 pounds per cubic foot and preferably not above about 5 pounds per cubic foot.

"The catalyst hydrocarbon feed suspension formed as above provided is passed through a riser contact zone for a hydrocarbon contact time less than about 5 seconds before discharge therefrom at a temperature sufficiently elevated to maximize recovery of vaporized hydrocarbon material separately from catalyst particles. In a more particular and specific aspect the present invention is directed to the catalytic conversion of high boiling residual oils comprising vacuum gas oils containing high boiling Conradson carbon producing materials employing a special ultrastable crystalline zeolite containing catalyst at a temperature equal to or above the feed pseudo-critical temperature in preferably a riser contact zone for a hydrocarbon residence time in the range of 0.5 to about 5 seconds and more usually not above about 3 seconds.

"Thus as the end boiling point of the hydrocarbon feed or the Conradson carbon level thereof increases so also will the catalyst regeneration temperature generally increased in response to increased deposited carbonaceous material removed by combustion and contributing to high temperature regeneration and conversion of the feed according to the concepts of this invention." [*Emphasis added.*]

<u>Claims 1 and 10:</u>

1. A method for .[.~~upgrading~~.]. *increasing conversion to liquid products of* a residual oil portion of crude oil boiling above 600°F. comprising metallo-organic compounds which comprises contacting *a feed containing* said residual portion of crude oil boiling above 600°F. *said feed having a Conradson carbon content above about 2.5 weight percent* with a catalyst consisting of from 20 to 80 wt. % of an ultrastable faujasite crystalline zeolite dispersed in a .[.~~silica-clay~~.]. matrix .[.~~for a time at a temperature particularly selective for conversion of the residual portion of crude oil to products of gasoline, light cycle oil and gasoline forming gaseous components; and~~
~~recovering said products comprising gasoline and light cycle oil~~.]. *selected from the group consisting of: silica-clay, silica-alumina, clay, silica, alumina and mixtures thereof, at a temperature above about 950°F. to provide a ratio of volume percent of light cycle oil to heavy cycle oil of in excess of 1.03 at essentially constant conversion and recover said light cycle oil and heavy cycle oil.* [*Emphasis in original.*]

10. The method of claim 1 wherein an atomizing diluent material is used with the residual oil feed comprising one or more materials selected from steam, CO2, light normally gaseous hydrocarbons comprising C3 minus materials in cooperation with atomizing spray nozzles.

The office action further concedes that *Dean* does not specifically disclose the feed has normal boiling points above 1000 °F, but asserts that it

would have been obvious to modify *Dean* "because one of skill in the art would use any feed having boiling points greater than 600 °F including a feed having a boiling point greater than 1000 °F with the expectation that any feed having boiling point greater than 600 °F can be treated in the process of Dean."

Significantly, the office action appears to overlook several key and critical differences between *Dean* and applicant's method, besides the boiling points of the feed. *Dean* is not a supercritical solvent process. In particular, *Dean* fails to teach or suggest (1) the use of solvating hydrocarbons, (2) a weight ratio of solvating hydrocarbons to feed hydrocarbons of at least 2:1, (3) operation of the reactor at a temperature above the critical temperature *of the feed/solvent mixture*, and (4) operation of the reactor at a pressure above the critical pressure *of the feed/solvent mixture*.

In claim 1, applicant recites that solvating hydrocarbons have a boiling point below 1000 °F and the feed hydrocarbons have boiling points above 1000 °F. *Dean's* purported disclosure of feed hydrocarbons boiling above 600 °F does not teach or suggest a mixture of feed hydrocarbons boiling above and solvating hydrocarbons below 1000 °F. Even if *Dean* did suggest feed hydrocarbons boiling above 1000 °F, it still would have failed

to suggest applicant's mixture of feed hydrocarbons boiling above 1000 °F with at least 2 parts by weight solvating hydrocarbons boiling below 1000 °F. Note that *Dean's* disclosure of an atomization diluent at 8/22-40 suggests dispersion of the feed oil as in droplets into a low density or gaseous phase to promote contact between the catalyst and oil droplet. This teaches squarely away from applicant's feed/solvent solution, where the solids are supplied merely as a heat source and physical or chemical contact between the feed hydrocarbon and (non-catalytic) solid particles is not required. Moreover, the *Dean* atomization diluent is steam, CO2, or a light normally gaseous hydrocarbon with 3 or fewer carbon atoms; steam and CO2 are not even hydrocarbons, and C3 minus gas is not a likely solvent at conventional near-atmospheric FCC pressures.

Claim 1 further recites that the weight ratio of low-boiling solvating hydrocarbons to high-boiling feed hydrocarbons is at least 2:1, i.e. no more than one-third feed hydrocarbons (excluding the recirculated solids). Applicant's claimed process requires the solvent to effect thermal conversion at supercritical conditions of the feed-solvent mixture. If the ratio of solvent to feed is too low, the supercritical temperature/pressure of the mixture are too high and/or thermal conversion is too low with excessive

coke make with reactor fouling or plugging. See the specification at paragraph and [0047].

Dean teaches squarely away from this approach, and instead relies on special ultrastable faujasite catalyst to inhibit coke make. In sharp contrast to applicant's claim 1, *Dean* discloses only that the optional atomizing diluent material, if it is even used, is present "in an amount which will reduce the high boiling feed partial pressure." In general, the partial pressure is understood by the skilled artisan to be proportional to the volume fraction of a vapor or gas, so that the presence of steam in the well known example of steam distillation, for instance, will result in more vaporization of a high boiling hydrocarbon. This suggests a lower operating pressure in *Dean* to facilitate vaporization of the feedstock, rather than the relatively high supercritical pressure condition in applicant's invention. *Dean* certainly does not preclude operation with a low solvent:feed ratio, and does not at all suggest the criticality of the 2:1 ratio claimed by applicant.

Claim 1 further recites that the reaction zone is maintained above the critical temperature of the feed mixture, i.e. the mixture of high-boiling feed hydrocarbons and solvating hydrocarbons. See paragraphs [0007] and [0041], *inter alia*. This is achieved by introducing the hot particulates and the feed/solvent mixture into the reaction zone so that the analogous thermal

equilibrium temperature (of both reactants and reaction products) is above the mixture critical temperature. The solvent serves to lower the critical temperature of the mixture as described in paragraph [0047] so that a desirably low operating temperature (relative to the feed mixture Tc) that is above the <u>mixture</u> critical temperature can be used. In contrast, *Dean* says only that *the regenerated catalyst* is at a temperature above 1350 °F and is equal to or above *the feed pseudo-critical temperature*; no teaching or suggestion is found anywhere in *Dean* to employ a relatively large solvent proportion (> 2:1) and a temperature throughout the reaction zone that is above the critical temperature of the mixture of feed and solvent (and reaction products).

Similarly, claim 1 recites that the reaction zone is maintained above the critical pressure of the feed mixture, i.e. the mixture of high-boiling feed hydrocarbons and solvating hydrocarbons rather than just the solvating hydrocarbons or the feed mixture. See paragraph [0041]. It is well known that the critical pressure of a hydrocarbon mixture generally tends to be higher than that of the individual hydrocarbon components. See paragraph [0074] and Figs. 8 -- 14 for several feedstock-solvent systems. *Dean*, in contrast, as noted above, operates under sub-critical conditions where vaporization of the feed occurs and the partial pressure is *reduced* to

facilitate vaporization. Nowhere does *Dean* provide any other teaching, suggestion or guidance as to the pressure to be employed in the FCC processing, let alone providing a supercritical pressure in the reaction zone. Note that typical FCC pressures presumably implied in *Dean* range from 0.15 to 0.2 MPa (22 to 29 psi), see *Kirk-Othmer Concise Encyclopedia of Chemical Technology*, pp. 856-857 (1985), copy attached, whereas critical pressures in claim 1 are generally orders of magnitude higher at 4.8 to 13.8 MPa (715-2015 psi) as noted in paragraph [0041].

Dean is an FCC process not at all similar to the present supercritical solution conversion process of the present invention, which is patentably distinct from *Dean*. See paragraph [0013]. It is respectfully requested that *Dean* be withdrawn as a section 103 reference.

Johnson and *Herbst* are cited as secondary references against claims 2-6 and 11-39. Since they are similarly directed to FCC processes, however, they similarly fail to fill the gap from the conventional FCC process of *Dean* to applicant's claimed invention. Neither reference teaches or suggests converting a high-boiling hydrocarbon with a hydrocarbon solvent at a solvent to feed ratio of at least 2:1 and at supercritical temperature and pressure of the feed-solvent mixture.

It is respectfully submitted that the present application is in condition for allowance. Should any issues remain that are appropriate to telephone resolution, please contact undersigned counsel.

Respectfully submitted,

Daniel N. Lundeen
Reg. No. 31,177
Lundeen & Dickinson, L.L.P.
P.O. Box 131144
Houston, Texas 77219-1144
(713) 652-2555
(713) 652-2556 Fax
ATTORNEY FOR APPLICANT